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HIGH ENERGY DENSITY FILM CAPACITORS (PREPRINT)

Shihai Zhang, Brian Zellers, Jim Henrish, Shawn Rockey, and Dean Anderson

Strategic Polymer Sciences, Inc.

Chen Zou and Qiming Zhang

The Pennsylvania State University

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Shihai Zhang, Brian Zellers, Jim Henrish, Shawn Rockey, and Dean Anderson
(Strategic Polymer Sciences, Inc.)
Chen Zou and Qiming Zhang (The Pennsylvania State University)

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Strategic Polymer Sciences, Inc.
200 Innovation Boulevard, Suite 237
State College, PA 16803

Penn State University
Materials Research Institute
University Park, PA 16802

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14. ABSTRACT
A series of high dielectric constant polymers have been developed with K from 10 to over 50. The high-K polymers have high dielectric breakdown strength above 700 V/µm and high energy density up to 27 J/cc in lab-scale small sample test. The capacitor film was produced by using melt-extrusion and biaxial orientation process and film thickness from 2 µm to 10 µm has been achieved. Metallized prototype capacitors show promising performance with energy density above 3 J/cc. Future development will target at 10 J/cc energy density in the packaged capacitors. Potential applications include pulsed power capacitors for medical defibrillators, high-power X-Ray/laser system, and direct energy weapon systems.

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Shihai Zhang ξ, Brian Zellers, Jim Henrish, Shawn Rockey, Dean Anderson
Strategic Polymer Sciences, Inc., 200 Innovation Blvd, State College, PA 16803, USA

Chen Zou, Qiming Zhang
Materials Research Institute, The Pennsylvania State University, University Park, PA 16802, USA

Abstract

A series of high dielectric constant polymers have been developed with K from 10 to over 50. The high-K polymers have high dielectric breakdown strength above 700 V/μm and high energy density up to 27 J/cc in lab-scale small sample test. The capacitor film was produced by using melt-extrusion and biaxial orientation process and film thickness from 2 μm to 10 μm has been achieved. Metallized prototype capacitors show promising performance with energy density above 3 J/cc. Future development will target at 10 J/cc energy density in the packaged capacitors. Potential applications include pulsed power capacitors for medical defibrillators, high-power X-Ray/laser system, and direct energy weapon systems.

I. INTRODUCTION

The commercial and consumer requirements for compact and more reliable electric power and electronic systems such as hybrid electric vehicles, microelectronics and medical defibrillators have grown substantially over the past decade. Capacitors are critical components in these devices for storing, filtering, controlling, and manipulating of electric charges. As a result, capacitors with high electric energy can enable the miniaturization of such electric and electronic devices.

Pulse-forming networks are critical for many pulsed power applications and they perform the conversion of prime electric energy into short pulses needed to energize loads that are required for directed energy and kinetic energy weapons and high power microwaves, electromagnetic armor, and megawatt-level uninterruptable power supplies. They require high energy density, fast discharge speed (milliseconds to nanoseconds) capacitors that cannot be fabricated with commercially available dielectric materials. Current commercial electrostatic capacitors usually have an energy density well below 2 J/cc due to the limitation of dielectric material performance and reliability constraints. The bulky size of capacitor components severely impedes the miniaturization of many electronic devices, although other active components have observed dramatic size reduction during the past 20 years. In some power electronic devices, capacitor components can occupy more than 50% of the device volume.[1]-[5]

The energy density (U_E) of a linear dielectric material is proportional to its dielectric constant (K) and the square of its dielectric breakdown strength (DBS) value. Therefore, high K and DBS values are critical to achieve a high energy density in capacitors. DBS is determined by both intrinsic and extrinsic (process-related) factors, but there is a general consensus that except for very thin films deposited on a substrate, the breakdown in practical components is limited by defects introduced during film processing. There have been many improvements over the last 10 to 20 years in energy storage capacitor fabrication so that energy densities of 1.5 to 3 J/cm^3 are now available. It is uncertain if more process improvements can be expected to yield further energy density increase, but there is little doubt the progress will be slow and evolutionary. Therefore, materials with significantly higher dielectric constant are required to achieve breakthrough in high energy density film capacitors.

In this report, we present our recent efforts in the development of high-K polymer dielectric materials, the commercial scale production of high-quality capacitor film, and the test of our first generation prototype capacitors.

II. HIGH-K POLYMER DIELECTRIC MATERIALS

Commercial polypropylene (PP) capacitor film has a K of 2.2 and is the most popular dielectric in film capacitor industry for more than 50 years.

There are several high-K polymeric dielectric materials been developed at Penn State University during the past 10 year.[6]-[15] The first is a high-energy electron irradiated Poly(vinylidene fluoride-co-trifluoroethylene) copolymer (PVDF-TrFE).[7] PVDF-TrFE is a classic ferroelectric polymer with Curie transition temperature (T_c) above 60 °C, depending on the composition. The irradiated PVDF-TrFE is converted to a relaxor ferroelectric material with the disappearance of the Curie transition and the giant dielectric constant above 50 at room temperature.

Similar high-K was also achieved by a chemical modification. A third monomer such as chlorotrifluoroethylene (CTFE) or 1-chloro-1-fluoro-ethylene (CFE) was introduced into PVDF-TrFE to obtain a terpolymer.[8] The CFE or CTFE is bulkier than VDF and TrFE and they convert the all trans beta crystalline

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ξ email: szhang@strategicpolymers.com

phase of PVDF-TrFE to trans-gauge alpha crystalline phase. Similar to the irradiated copolymer, the terpolymers are also relaxor ferroelectric materials with K >50 at room temperature. The terpolymers are thermoplastic and can be processed into thin capacitor film by either solvent cast or melt extrusion/orientation.

The dielectric constant of the high-K polar fluoropolymers was further improved by the development of nanocomposites.[13]-[15] With well-controlled morphology, dielectric constant over 1,000 has been achieved in PVDF-TrFE-CFE/CuPc nanocomposites.

Other PVDF-based copolymers have also been evaluated by us for high-energy density film capacitor applications since 2005.[6],[9]-[12] It has been found that the copolymers have significantly better performance than the PVDF homopolymer. Figure 1 presents the dielectric constant of these high-K polymers at 1 kHz as a function of temperature.

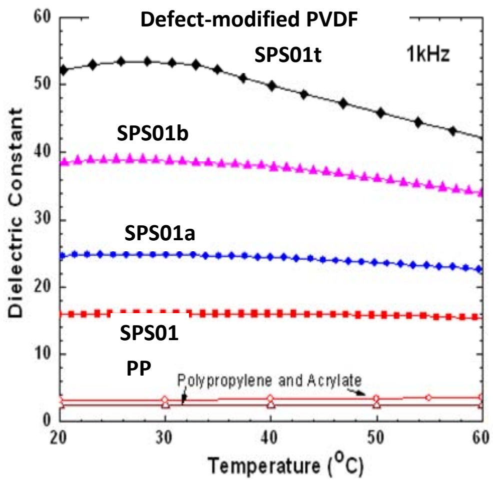

Figure 1. Dielectric constant of SPS high-K polymers at 1 kHz. They are purely thermoplastic polymers without any filler.

While the dielectric constant of these modified PVDF is high, they also have high dielectric breakdown strength for high energy density applications. Figure 2 compares the dielectric breakdown strength of SPS high-K polymer and the commercial 4.8 μm thick PP capacitor film. The former was produced with melt extrusion and biaxial orientation. It has a thickness of 10 μm.

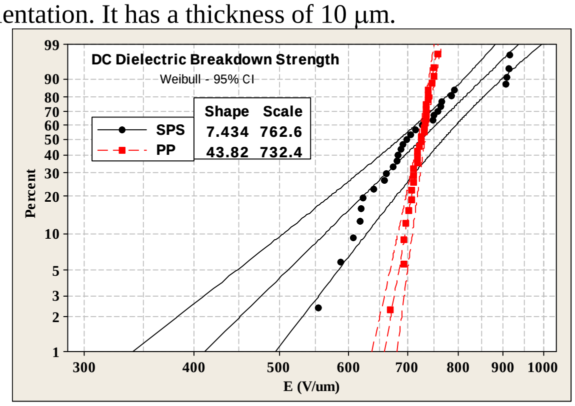

Figure 2. Comparing the DC dielectric breakdown strength of SPS high-K capacitor film with commercial PP capacitor film. Lab-scale sample area ~1 cm^2.

It can be seen that the SPS capacitor film has similar Weibull dielectric breakdown strength as commercial PP capacitor film. At lab-scale film, both have breakdown strength over 700 V/μm. However, the commercial PP film has a Weibull shape parameter over 43, while it is only 7.4 for SPS capacitor film. This indicates that SPS capacitor film quality is not as good as the commercial PP film and the dielectric breakdown strength in large capacitors of SPS film will be significantly lower than commercial PP.[16]-[20] For example, there are over 20% of SPS samples failed below 650 V/μm, while none of PP sample failed.

On the other hand, the high Weibull breakdown strength does suggest that SPS capacitor film has high intrinsic breakdown strength, and the Weibull distribution should be able to be improved with cleaner resin and better film processing control.

With the high dielectric breakdown strength and high dielectric constant, the SPS capacitor film has significantly higher energy density than commercial PP capacitor film. Figure 3 compares the discharged energy density of SPS film and PP film based on the test of ~1 cm^2 sample size.

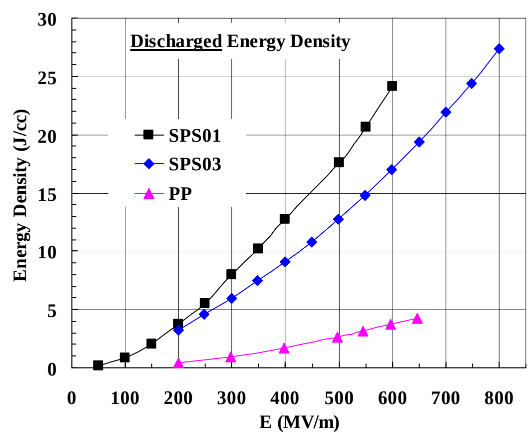

Figure 3. Discharged energy density of SPS high-K film and commercial PP film as a function of applied electric field. The data are directly measured values and not theoretical prediction.

It can be seen that energy density of 27 J/cc can be obtained in SPS03. At 500 V/μm, SPS01, SPS03, and PP have energy density of 17.6, 12.8, and 2.7 J/cc, respectively.

III. PRODUCTION OF CAPACITOR FILM

While high dielectric breakdown strength and energy density have been achieved in lab-scale capacitor film samples, it is critical to scale up the capacitor film production for the development of prototype capacitors.

We have performed pilot-scale capacitor film production using melt-extrusion biaxial orientation process. The pilot film line can produce capacitor film up to 2 m wide and run at over 200 m/min. Capacitor film rolls with thickness from 2 μm to 10 μm have been successfully produced. The length of the pilot film roll is over 3,000 m and the thickness uniformity is better than ±5%. High dielectric breakdown strength is confirmed again in the large capacitor film rolls.

Figure 4 shows a picture of SPS capacitor film rolls.

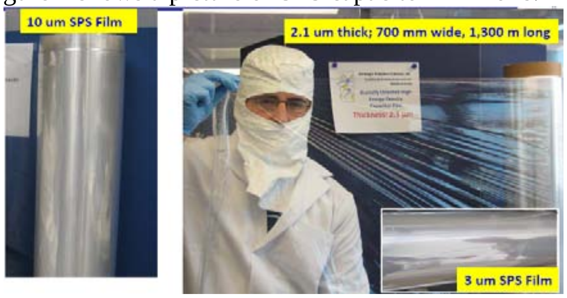

Figure 4. High quality ultra-thin SPS capacitor film produced by melt-extrusion and biaxial orientation.

It should be noted that the melt extrusion process is widely used by capacitor film industry to produce PP and PET capacitor film. Commercial capacitor film line can produce film with thickness below 1 μm thick, above 8 m wide, and at a speed faster than 200 m/min. The large scale solvent-free production can significantly reduce the capacitor film cost as compared with the solvent-based capacitor film production, in addition to the environmental advantage.

IV. TEST OF PROTOTYPE CAPACITORS

The film was metallized and wound into prototype capacitors with different capacitances. The metallization is a thin layer of aluminum with high surface resistance for high dielectric breakdown strength.

10 μF capacitors made with 5 μm thick film exhibit high dielectric breakdown strength and high energy density. Most of the 10 μF capacitors can pass 1,500 V breakdown test, and more than half of them can survive 2,000 V charge-discharge test. The capacitor was charged under constant current to the preset voltage, and then discharged to a load resistor. The discharged energy density was calculated from the discharging process by numerical integration.

Figure 5 shows pictures of the prototype capacitors. The 85 uF capacitors were made with 3.1 μm thick SPS high-K film. It has capacitance density of 5.72 uF/cc or 3.13 uF/g. For comparison, commercial PP capacitors with 3 μm thick film have capacitance density of 0.62 uF/cc or 0.45 uF/g. This clearly demonstrates the potential miniaturization of many electric devices by using SPS high-K film capacitors.

Figure 5. Prototype high energy density film capacitors.

A. High Energy Density

Figure 6 presents the discharged energy density as a function of applied electric field.

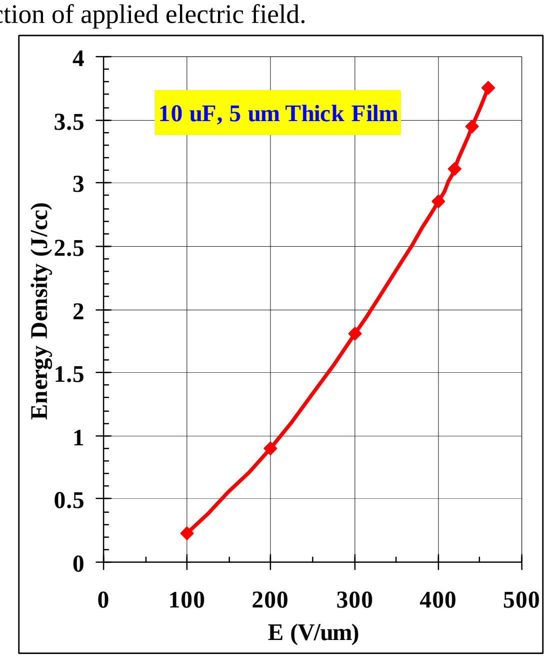

Figure 6. Discharged energy density of SPS high-K prototype capacitors made with 5 μm thick film..

The 10 μF prototype capacitors have an energy density up to 3.75 J/cc at 460 V/μm. It should be noted that the only 37% of the 10 μF capacitors is active capacitor film while the other 63% is inactive edges and winding core. At the film level, the capacitor has energy density about 10 J/cc. The winding volumetric efficiency can be improved by a better capacitor design.

B. Millisecond Discharge

Figure 7 presents the discharge behavior of an 87 μF capacitor with 3.1 μm thick film to a 50 Ω load resistor.

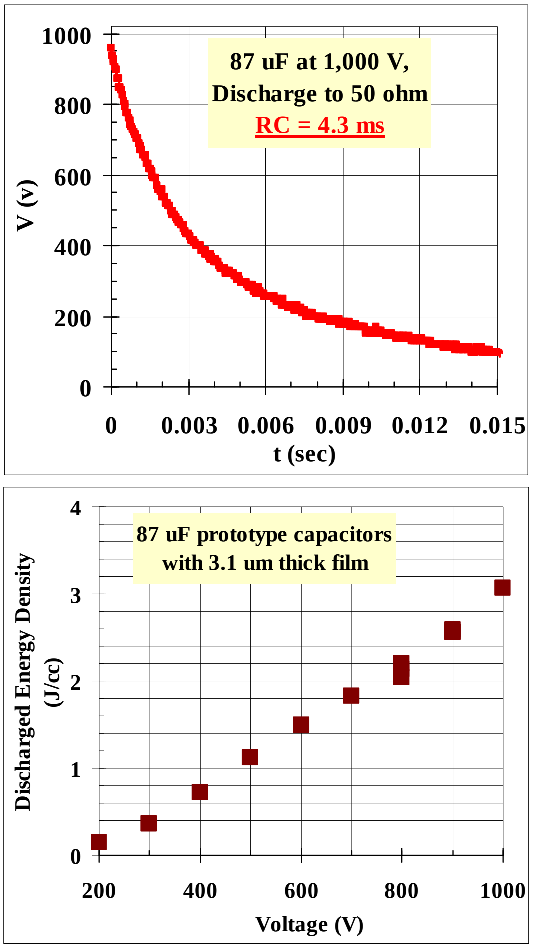

Figure 7. Discharged energy density of SPS high-K prototype capacitors made with 3.1 μm thick film..

It is clear that the 87 μF capacitor can discharge at millisecond time scale.

As compared with the 10 μF 5 μm prototype capacitor, the 87 μF capacitor has higher volumetric packaging efficiency of 57%, but the film is thinner and film area is larger. Therefore, its dielectric breakdown strength is lower than the 10 μF prototype capacitors at this time.

V. FUTURE PLAN

While the high energy density of SPS polymer high-K materials has been demonstrated in lab-scale small area capacitor film samples and the pilot production of ultra-thin capacitor film has been performed with inexpensive melt extrusion biaxial orientation process, it is clear that the capacitor film quality still needs significant improvement to achieve the high energy density as measured in the lab-scale film sample. Specifically, 1 cm^2 size lab-scale capacitor film has dielectric breakdown strength above 750 V/μm (comparable to PP film) and energy density above 27 J/cc, the 10 μF prototype capacitors have breakdown voltage well below 500 V/μm. It is believed that the contamination in the resin and the film significantly reduces the dielectric breakdown strength and the energy density in the large prototype capacitors. Therefore, it is critical to produce capacitor resin and film with quality comparable to commercial PP capacitor film to fully transfer the high energy density from lab-scale small area film sample to large size capacitor devices.

Once the film quality of the high-K polymers has been improved and the prototype capacitors can be operated at 600 V/μm, energy density above 10 J/cc can be achieved by combining more efficiency capacitor design.

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